INTERACTION OF SURFACTANTS WITH SHEAR FLOWS AND SURFACE WAVES

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LONG-TERM GOAL

The long-term objective is to develop noninvasive instrumentation and techniques for in situ measurements of the interfacial properties of the ocean, including the *intrinsic* viscoelastic properties. These intrinsic properties, as interpreted through the laws of mechanics, will be related to the apparent properties of the interface as deduced from the usual phenomenological approach of short-wave dampening. Ultimately, the intrinsic interfacial properties are required for computational models of the ocean surface, derived from first principals, to predict the shape, motion and other factors that influence radar scatter from a surfactant-influenced interface such as the ocean surface.

The rapid increase in computing power in the past two decades has made it possible to numerically solve the exact equations of fluid motion over a moderate range of scales in time and space. And although the equations of motion for the flow in the air and in the sea, namely the Navier-Stokes equations, have been known for over a century and a half, the boundary conditions for these equations are generally unknown at a surfactant-influenced gas/liquid interface because the boundary conditions depend on the intrinsic (cf apparent) viscoelastic properties of the interface, measurements of which were not feasible until recently

SCIENTIFIC OBJECTIVES

The scientific objective of the present project is to determine the intrinsic viscoelastic properties that have a significant effect on the movement, shape, and other factors that influence radar scatter from a surfactant-influenced gas/liquid interface such as the ocean surface.

Initially, through the use of the most general stress-strain relation for a Newtonian interface, namely the Boussinesq-Scriven constitutive relation, we wish to directly determine the viscoelastic properties of the interface. This must first be performed in the laboratory for well-behaved surfactant systems and well-controlled subsurface flows and waves, prior to the application of the technique to in situ measurements. Ultimately, non-Newtonian constitutive relations will be utilized for the evaluation of the viscoelastic properties of natural polymeric films on the ocean surface.

There are two coefficients in the Boussinesq-Scriven that describe the dissipative or the viscous part of the stress at the interface. These are the interfacial shear viscosity (μ^S) and interfacial dilatational viscosity (κ^S). In general, these coefficients depend on the thermodynamic state of the surfactant-influenced interface, i.e. the surface concentration of the surfactant. In addition to the viscous effect at surface, the elasticity of the surface, which results from gradients in the

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thermodynamic surface tension must be quantified. The fluid motion adjacent to the interface is coupled to the interface due to the viscosity in the bulk of the fluid. Thus, in order to quantify the viscoelastic effect at the interface, the bulk fluid motion adjacent to the interface as well as the composition and motion of the interface itself must be measured. Reproducible and consistent measurements of the intrinsic viscosities, especially κ^s are yet to appear in the literature for any surfactant system. Published measurements of κ^s for a given surfactant vary by as much as a factor of 10^5, depending on the method used to measure κ^s.

APPROACH

Well-controlled flow fields are being utilized in the laboratory in order to study the viscoelastic properties of a water surface covered by various monolayers. The unsteady dilation and compression of the ocean surface resulting from atmospheric turbulence (wind), shear in the liquid side and surface or internal waves is simulated in the laboratory by the flow field due to the rise and consequent interaction of a laminar vortex pair (see Figure 1). The vortex pair stretches the surface without intrusive barriers, such as in Langmuir trough-type of devices. Also, we have recently developed other flow facilities including a c-g wave tank system as well as a steady-state flow loop with a film under compression by a uniform subsurface flow (“Reynolds ridge device”) designed primarily as a benchmark test for the new areal SHG system. The boundary layer beneath the surface is examined in each system. The c-g waves are used as an extension of the vortex pair study in order to stretch and dilate the surface, thus allowing the measurement of the viscoelastic properties of the surface. The c-g waves are also used to study the relationship between the intrinsic viscoelastic properties and wave dampening characteristics, which affect the radar back scatter from the ocean surface.

The velocity measurements in the bulk of the fluid must include the free surface boundary layer, which for high Reynolds number flows can be very thin (of the order of a millimeter or less). Thus, the noninvasive laser-based method of digital particle image velocimetry (DPIV) is undergoing further refinement in our laboratory to improve the spatial and temporal resolution of the velocity field measurements. In order to compliment the surface viscosity measurement techniques that we have developed, a deep-channel surface viscometer is also being tested. The deep-channel viscometer provides measurements of the interfacial shear viscosity (μ^s) which can be subtracted from the measured value of the sum of the surface viscosities to provide κ^s.

WORK COMPLETED

Many tasks have been accomplished in the first year of the project. A new laboratory has been established. The new laboratory is located in a building with an underground connection to Prof. Korenowski's laboratory. This has permitted safe transportation of delicate equipment, including the high frequency (100 Hz) Infinity Laser and support systems, to and from each laboratory without exposure to the elements. Our new laboratory has been equipped with a high capacity water distillation system and air filtration facilities. A large clean-water tank with a 600 liter capacity for vortex measurements has been put into operation. The deep-channel viscometer was completed and put into operation.

A Reynolds ridge device (see Figure 2) was constructed of inert materials (glass, acrylic plastic, and stainless steel) and is being used initially as a tool to tests and calibrate the areal SHG system in flowing systems. Since the strain and the resulting stress in the boundary layer beneath the film can be measured directly with laser velocimetry, the surfactant concentration gradient on the surface can be determined and used for evaluating the concentration profile measured by the SHG camera.

RESULTS

The instrumentation for noninvasive probing of surfactant films in the laboratory (via SHG and DPIV) was developed and the results have been published (Hirsa, et al. 1997a). Our measurements provide data on the evolution of the surfactant concentration field during the interaction of a transient subsurface flow. The analysis presented in that paper showed, for the first time, that quantitative information on the flow in the bulk fluid can be directly obtained from nonlinear optical probing of the surface composition. Also, a novel technique for the direct determination of the sum of the surface dilatational and shear viscosities ($\kappa^S + \mu^S$) using SHG and DPIV data was developed for insoluble surfactants and the results were published (Hirsa, et al. 1997b). The extension of this technique to soluble systems, and ultimately to natural ocean films is the central thrust of the present effort and is expected to become feasible once the areal SHG system is fully tested. Toward that end, a Reynolds ridge device was built and tested and the results follow.

A new flow loop was specially designed and constructed to study surfactant films under steady-state compression and to provide a benchmark test of the areal SHG system in flowing systems (see Figure 2). An existing Laser-Doppler Velocimeter (LDV) system has been modified to provide scanning velocity measurements with high spatial resolution (50 microns) simultaneously with the areal SHG measurements. The LDV measurements (i.e. pointwise measurements made relatively quickly along the surface-normal direction) provide the instantaneous shear stress on the film which must be balanced by the gradient in the surface tension and hence surfactant concentration. Preliminary LDV measurements of the shear stress beneath a hemicyanine monolayer film, made in the new flow loop, are presented in Figure 3, along with comparisons to the laminar boundary layer theory on a solid wall. Figure 3 shows that although the stress distribution beneath the surface film is linear when plotted with the appropriate scaling in agreement with the theory for a solid wall; interestingly, the stress is greater than the stress beneath a solid wall. The SHG data that was acquired simultaneously along with the above LDV measurements are expected to be analyzed in the next few days and will be presented at the APS meeting in San Francisco (Korenowski, Hirsa, Van Wagenen & Jin 1997).

A novel technique has been developed and successfully tested which provides the instantaneous shape of the air/water interface, thus permitting the DPIV measurements to be performed in *boundary-fitted* coordinates (i.e., measurements on grids fitted to the deforming air/water interface instead of orthonormal coordinates used in conventional DPIV). In the boundary-fitted DPIV technique, the particle images are first utilized to map the location of the surface through an robust, automated cross-correlation method. This is possible because of the total internal reflection that occurs when viewing submerged objects from below the surface. Surface elevations of up to 2 mm at any x-position can thus be determined to an accuracy of better than 10 microns. This technique provides a two-dimensional mapping of the surface. The boundary-fitted measurements are particularly important because of the large gradients that exist in the velocity within the free-surface boundary layer. The results of this work will be presented at the

APS meeting (Mortzheim, Gayton & Hirsa 1997). Representative measurements with the boundary fitted DPIV are presented in Figure 4. The variation of the surface location η (averaged over the 4 mm field of view) as a function of time is shown in Figure 4(a). Figure 4 shows the surface upwelling due to the arrival of the vortex pair. The surface deformations associated with the secondary vortex structure is also evident in Figure 4. The surface velocity us is presented in Figure 4(b) and its surface-normal gradient, i.e. the surface viscosity, is shown in Figure 4(c). Figure 4 shows that the vortex pair flow field dilates the surface until the surfactant is completely cleared away from the center for a period, this process continues until the relatively larger surface tension of the clean region pulls back the film toward the center.

IMPACT/APPLICATION

We have shown for the first time that the intrinsic viscoelastic properties of surfactant-influenced surfaces may be measured directly through optical techniques. These intrinsic properties are required in numerical simulations of fluid motion on each side of the air/sea interface.

TRANSITIONS

We expect to develop the instrumentation and techniques to perform boundary-fitted DPIV measurements in the free surface boundary layer beneath ambient waves on the ocean within the next two years; this will compliment the nonlinear optical measurements of the surface.

RELATED PROJECTS

As described throughout this report, the present research project is closely linked to the research in nonlinear optical probing of the ocean surface, conducted by Prof. Korenowski in the Chemistry Department at RPI. Our synergistic approach functions in such a way that their efforts will continue to provide us with the complementary tools needed for measurements of the viscoelastic interfacial properties in the laboratory and ultimately in situ, and our flow facilities and flow measurements provide the benchmark for the development and testing of their surface probes prior to their implementation for in situ studies.

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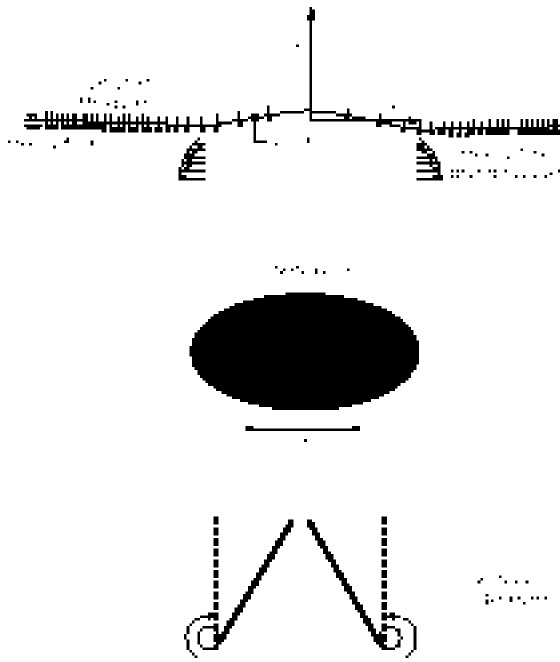


Figure 1. Schematic Diagram of the Generation and Interaction of a Vortex Pair with a Surfactant-Influenced Free-Surface.

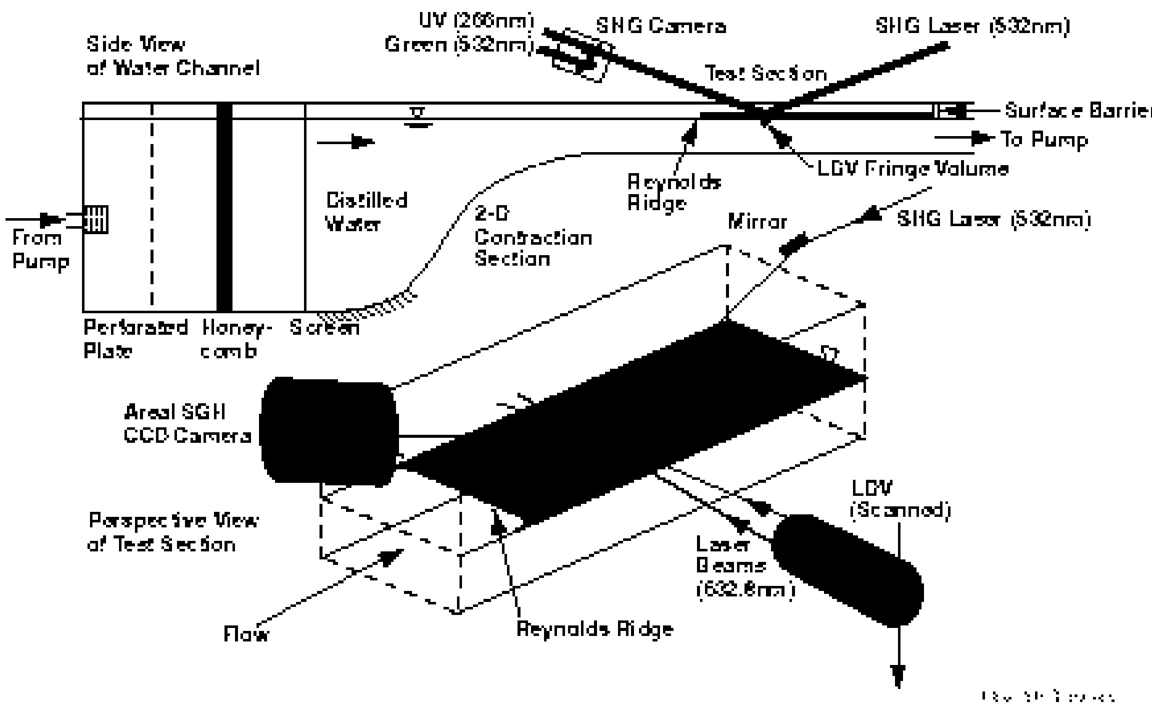


Figure 2. Schematic Diagram of the Flow Loop used for Calibration of the Areal SHG system. Note: The Reynolds Ridge maks the leading edge of the Surfactant film.

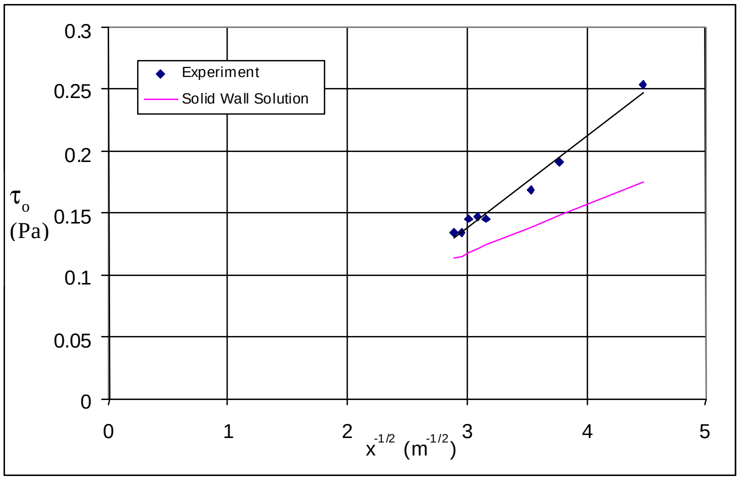
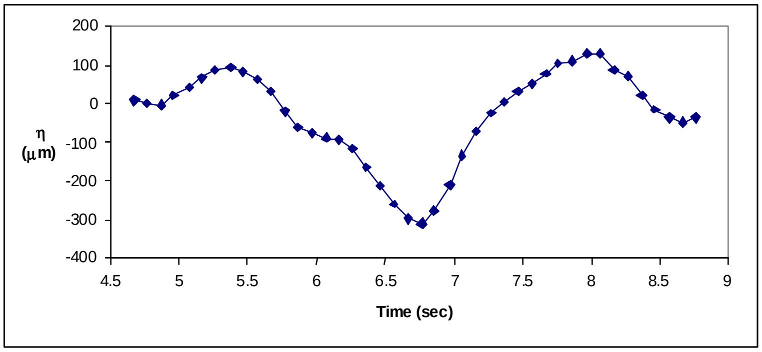
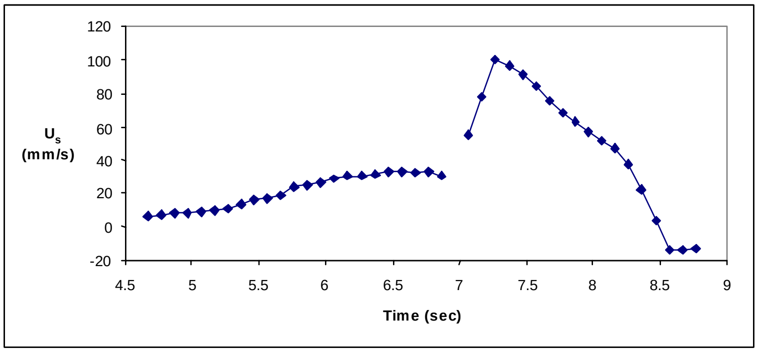


Figure 3. Shear Stress Beneath Surface Film, τ_0 as Measured by the LDV.
Note: The solid wall solution is due to Blausius.



(a)



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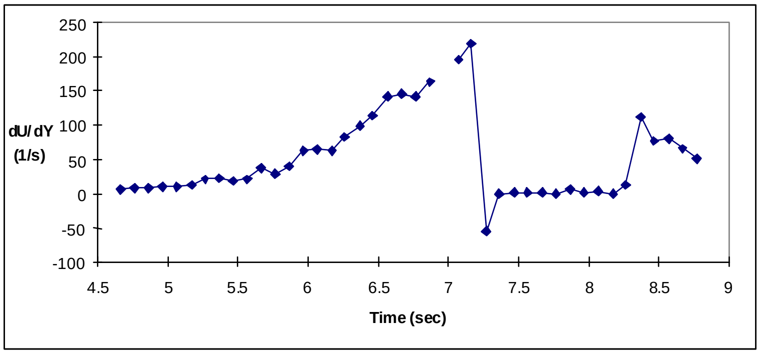

(c)

Figure 4. Representative measurements with the boundary-fitted DPIV technique.

It is showing the [spatially-averaged] surface location, h, as a function of time (a); interfacial velocity, u_s (b); and [surface-normal] shearing strain rate [= surface vorticity] du/dy (c). These measurements were made at x = 4 cm and π = 3.2 dynes/cm.